ULTRAFAST FREQUENCY SHIFTING BY ABSORPTION SATURATION OF QUANTUM-WELL EXCITONS IN A PHOTONIC-BAND STRUCTURE

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ABSTRACT

Transient spectral modulation associated with excitonic absorption saturation in a semiconductor quantum-well photonic-band structure is characterised by femtosecond transient reflectance spectroscopy. A large frequency modulation in a bandwidth up to 1 THz is observed for the absorption saturation of quantum-well excitons in the photonic-band structure at wavelengths near the resonance with the lowest heavy-hole excitons. Such a large frequency modulation near the peak of the exciton resonance can not be explained by refractive index change, but by Doppler frequency shifting caused by the ultrafast movement of pulse reflection wavefront in the photonic-band structure.

INTRODUCTION

Exciton is hydrogen-like bound state of electron-hole pair in semiconductors [1]. Two-dimensional excitons in semiconductor quantum wells exhibit large ultrafast optical nonlinearity associated with absorption saturation at room temperature [2]. Room temperature ultrafast absorption saturation of quantum-well excitons is interested because of its importance in applications to ultrafast photonic devices such as semiconductor mode-locked lasers and all-optical modulators [3]. Ultrafast absorption saturation of quantum-well excitons has been extensively studied under resonant excitation in pump-probe transient absorption measurements using femtosecond optical pulses. Femtosecond response in the probe intensity change has been observed and analysed in terms of exciton dynamics at room temperature [4, 5]. Spectral modulation in the process of the ultrafast absorption saturation, on the other hand, has not been reported so far at wavelengths near the exciton resonance. This is because that no change in refractive index occurs at the peak of exciton absorption line, as shown in exciton density dependence of refractive index spectra obtained from Kramers-Kronig transformation of unsaturated and saturated exciton absorption spectra [2]. Temporal change in refractive index was usually considered as the main origin of transient optical phase shift, and hence no spectral modulation was expected for ultrafast absorption saturation of quantum-well excitons at wavelengths near the exciton resonance [6].
In this paper, we show, however, that significant spectral modulation is produced during ultrafast absorption saturation of quantum-well excitons in a planer photonic-band structure consisting of periodic bi-layers. Spectral modulation associated with absorption saturation of quantum well excitons extends over a optical frequency range of nearly 1 THz. Ultrafast spectral modulation is mediated by exciton-induced self-phase modulation in the photonic-band structure. The origin of the excitonic self-phase modulation is not by refractive index change but by change in light propagation length of reflected light from the photonic-band structure. The light propagation length is limited by average reflection front defined for the mode profile of the optical waves confined in the photonic-band structure. Average reflection front moves in femtosecond time scale during ultrafast absorption saturation of the excitons. Doppler frequency shifting thus occurs for ultrashort optical pulses reflected from the photonic-band structure [7]. Large and ultrafast absorption saturation of quantum-well excitons in the photonic-band structure leads to high-efficiency frequency modulation of ultrashort optical pulses and is useful to optical pulse shaping and control in ultrafast photonics. An example of application of the exciton-induced Doppler frequency shifting is presented as optical pulse compression using the photonic-band structure.

NONLINEAR PHOTONIC-BAND STRUCTURE

The photonic-band structure was a multi-layer structure, of which constituents were grown on an undoped InP substrate by organo-metallic vapour phase epitaxy [8]. The photonic-band structure was formed of 20 unit cells, each of which consisted of 10-nm InP, 7-nm InGaAs quantum well, 100-nm InP and 109-nm InGaAsP in the order from the upper to the lower layers on the substrate. The structure is illustrated in Fig. 1. Each unit cell act as a Bragg layer with Bragg resonance wavelength of 1500 nm. The resonance of the lowest heavy-hole excitons occurs at a wavelength of 1500 nm for 7-nm InGaAs. Dual resonance of excitons and photons thus appears at 1500 nm in the photonic-band structure.
Photons are accumulated in the InP/InGaAsP bi-layers due to constructive interference of the optical waves inside the structure. Optical intensity has an maximum at a position in each quantum well and photon-exciton interaction is

enhanced in the photonic-band structure. The photon accumulation leads to the enhancement of linear optical absorption and absorption saturation [8].

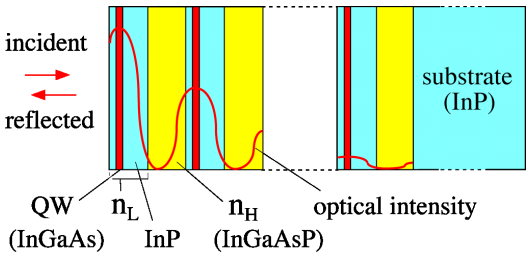

Fig. 1. Cross-section of the nonlinear photonic-band structure

TRANSIENT REFLECTANCE SPECTROSCOPY

Transient reflectance measurements were performed for the photonic-band structure at room temperature using the optical pulses having 180-fs duration, 17-nm spectral width and 1500-nm peak wavelength for the pump and probe pulses. Change in the magnitude of transient reflectance was limited by optical absorption and confirmed to be caused by ultrafast absorption saturation of the quantum-well excitons. Transient absorption trace is presented in Fig. 2 (a) with pump-probe correlation trace. Pump and probe pulses were identical, and the correlation trace reflected the pulse shape from the laser. The absorption was saturated instantaneously under resonant exciton creation by the pump pulses at zero delay time. The absorption was then recovered with a time constant of 200 fs. The ultrafast absorption recovery was produced by exciton dissociation via collision with optical phonons. At the peak of the transient absorption saturation, nearly 50 % change in the absorption was produced due to the enhanced photon-exciton interaction in the photonic-band structure.

Frequency-resolved transient reflectance measurements were performed to characterise spectral modulation during the process of the excitonic absorption saturation in the photonic-band structure [8]. The reflectance spectroscopy measurements provided transient reflectance spectrograms as shown in Fig. 2 (b). Change in the transient reflectance was plotted in pseudo-colours and normalised with its maximum. Black dots superimposed on the reflectance spectrogram represent the average wavelength of transient reflectance spectrum at each step of the delay time. The average wavelength was obtained using the reflectance spectrogram as a distribution function.

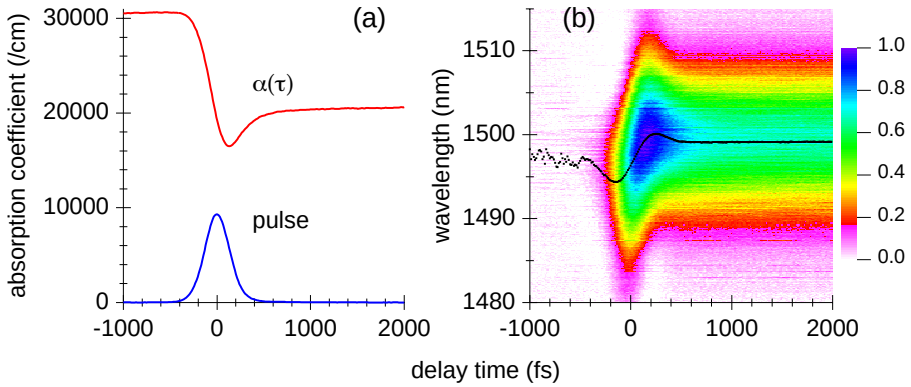

Fig. 2. (a): absorption $\alpha(\tau)$ and pulse correlation traces. (b): transient reflectance spectrogram.

Average optical frequency of the transient reflectance spectrogram was calculated from the average wavelength in Fig. 2 (b) and plotted as a function of delay time in Fig. 3. The time derivative of normalised transient absorption coefficient, which is written as $d\{\alpha(\tau)/\alpha_{max}\}/d\tau$, is also presented in Fig. 3. Here, α_{max} indicates the maximum unsaturated absorption coefficient. The average optical frequency and the time derivative of the normalised absorption are similar in their temporal shapes. The average frequency is thus proportional to the time derivative of the absorption coefficient with a constant of - 1.7. The constant is shown to represents linewidth enhancement factor for the quantum-well excitons in the photonic-band structure on the basis of rate-equation model [9]. For unbound electrons and holes which show broad and flat optical absorption spectrum, linewidth enhancement in transient absorption saturation has been well explained by theoretical refractive index change associated with spectral hole burning [10]. For excitons, on the other hand, absorption spectrum has a much narrower linewidth than that for the unbound states, and the linewidth enhancement factor of excitons is negligibly small. The linewidth enhancement factor of the quantum well excitons was

calculated from theoretical refractive index change to be – 0.06, almost 1/30 of the value experimentally obtained from the proportionality between the average frequency and the time derivative of the transient absorption. The small theoretical linewidth factor results from the fact that refractive index change is zero at the peak of a narrow absorption line [6].

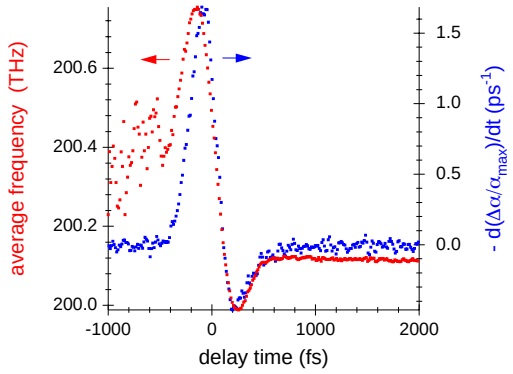

Fig. 3. Average frequency of transient reflectance spectrogram (red) and time derivative of $\alpha(\tau)$ (blue).

However, spectral modulation is possible in the photonic-band structure via another mechanism independent of refractive index change. Light propagation length before reflection is limited by average reflection front defined from the intensity profile of the confined optical mode in the photonic-band structure. The intensity profile depends on the magnitude of absorption saturation and the position of the average reflection front moves according to temporal evolution of the excitonic absorption saturation. From steady state transfer matrix calculation including exciton-density dependent absorption coefficient, it was confirmed that the average reflection front moves closer toward the surface of the photonic-band structure when the magnitude of the absorption saturation is increased. Ultrahigh speed movement of the average reflection front produces Doppler frequency shift of the reflected pulse spectrum. The amount of Doppler frequency shift was estimated from change in theoretical group delay time for the reflected wave. Change in the group delay time obtained to be a few femtoseconds. This produces an optical frequency shift in the order of 1 THz. Optical Doppler effect thus plays a significant role in the spectral characteristics of ultrafast absorption saturation of excitons in the photonic-band structure.

OPTICAL PULSE COMPRESSION

The spectral modulation by the Doppler effect is the origin of the self-phase modulation in the photonic-band structure. The self-phase modulation produced spectral broadening and pulse chirping, and allows optical pulse compression when pulse chirping is compensated. The Doppler frequency shifting in the photonic-band structure is thus useful for optical pulse compression. Femtosecond optical pulses incident to the photonic-band structure induced the self-phase modulation, and reflected optical pulses were spectrally broadened and chirped as shown in the optical pulse spectrograms in Fig. 4. The reflected optical pulses were negatively chirped: optical intensity in the reflected pulses was distributed from the shorter wavelengths to the longer wavelengths. The negative chirping was compensated by a positive chirping of the same absolute magnitude, as shown for the compressed pulse spectrogram in Fig. 4. The positive chirping was produced at a dispersive optical delay line [11]. The autocorrelation traces obtained from the spectrograms provided 380 fs and 270 fs widths before and after the pulse compression. A compression factor of 1.4 was thus obtained in the optical pulse compression using the photonic-band structure.

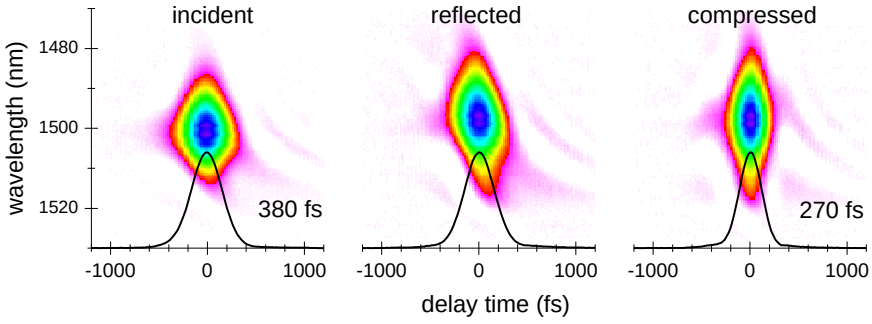

Fig. 4. Spectrograms of incident, reflected and compressed pulses with autocorrelation traces.

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